



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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(54) Title: METHOD FOR THE CONTROL OF VEGETATION USING HERBICIDAL COMPOSITION CONTAINING CARBOXYLIC OR PHOSPHONIC ACID SALT			
(57) Abstract			A method for selectively controlling undesirable vegetation by contacting the leaves with a herbicidal composition comprising a carboxylic or phosphonic acid salt known to form a coordination compound with calcium or magnesium salts. In a preferred embodiment, the carboxylic or phosphonic acid salt is an alkali salt of at least one acid selected from an aminopolycarboxylic acid, an aliphatic carboxylic acid, a hydroxycarboxylic acid, an amino acid, an ether polycarboxylic acid, a phosphonic acid and a polyphosphonic acid.

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TITLE: METHOD FOR THE CONTROL OF VEGETATION USING HERBICIDAL COMPOSITION CONTAINING CARBOXYLIC OR PHOSPHONIC ACID SALT

SPECIFICATION

Field of the Invention

The present invention relates to the use of a selective herbicidal composition containing a carboxylic or phosphonic acid salt.

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Background of the Invention

Chemicals used to kill plants can be classified as either non-selective herbicides or selective herbicides. Non-selective herbicides kill all plants to which they are applied. An example of a non-selective herbicide is N-(phosphonomethyl) glycine, known commonly as glyphosphate or "Roundup®." A selective herbicide kills only certain plants, that is, it is more toxic to some plants than to others. To be of practical use, a selective herbicide must have the ability to kill undesirable plants without affecting the remaining desirable plants. An example of such a chemical is the well known herbicide 2,4-dichlorophenoxyacetic acid (2,4-D) which will selectively kill broad-leaved weeds in, for example, turf grass. However, there are many weeds which are not destroyed by 2,4-D. Furthermore, 2,4-D is toxic and can cause eye irritation and gastrointestinal disturbances if ingested. Some undesirable grasses, like crabgrass or bermudagrass, can be selectively removed from turf by the use of arsenic compounds, but these herbicides can be toxic to both humans and animals. Other turf weeds, such as nimblewill, are not affected by 2,4-D or arsenic compounds and can only be controlled chemically by using

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glyphosphate which kills not only the nimblewill but all of the desirable plants surrounding the nimblewill.

U.S. Patent 5,668,086, herein incorporated by reference, discloses the use of a herbicidal composition containing diquat (1,1'-ethylene-2,2'-bipyridinium dibromide) or paraquat (1,1'-dimethyl-4,4'-bipyridinium dichloride) as contact herbicides. Diquat and paraquat have no selectivity but exhibit strong week-killing power by foliage treatment. In use, they are diluted with water. An ionic surfactant is further added so that the liquid can be applied evenly and ensure contact with the stems and leaves of the vegetation long enough to penetrate into the plant body. Unfortunately, the addition of the surfactant lowers the herbicidal effects of the composition. The reduction observed in the herbicidal effects observed in the composition is dependent upon the kind and amount of anionic surfactant used.

It is the primary object of this invention to provide a method for the selective control of certain undesirable vegetation using herbicides which are non-toxic to humans, animals and insects. It is a further object to provide such herbicides that also are odorless, water soluble and biodegradable.

Summary of the Invention

Undesired vegetation may be killed by wetting the leaves of the vegetation with a herbicidally effective amount of a composition comprising water and a carboxylic or phosphonic acid salt known to form coordination compounds with calcium or magnesium ions. Coordination compounds are substances with chemical structures in which a central metal atom such as calcium is surrounded by groups of non-metallic atoms joined to it by chemical bonds.

Examples of the salts of acids which form complexes with calcium or magnesium ions and are useful in the invention are salts of at least one carboxylic or phosphonic acid selected from aminopolycarboxylic acids, aliphatic carboxylic acids, hydroxycarboxylic acids, amino acids, ether polycarboxylic acids, phosphonic acids and polyphosphonic acids. The compositions of the invention act as selective herbicides. The composition may further contain a surfactant as well as other conventional adjuvants.

Since the mode of action appears to be through the leaves of the vegetation, there is little, if any, residual herbicidal effect in the ground. Thus, it is possible to grow desirable plants adjacent to and around the treated area.

Detailed Description of Preferred Embodiments

5 Unwanted vegetation may be killed by wetting the locus of the vegetation with an aqueous composition containing a carboxylic or phosphonic acid salt known to form coordination compounds with calcium or magnesium ions. In a preferred embodiment, the invention relates to a herbicidal composition containing, as the carboxylic acid salt, the salt of carboxylic or phosphonic acids selected from aminopolycarboxylic acids, aliphatic carboxylic acids, hydroxycarboxylic acids, amino acids, ether polycarboxylic acids, phosphonic acids and polyphosphonic acids.

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15 The carboxylic and phosphonic acids for use in the composition include aminopolycarboxylic acids having from 1 to about 10 carbon groups, a C₁-C₈ aliphatic carboxylic acid, a C₁-C₈ hydroxycarboxylic acid, an amino acid and ether polycarboxylic acids having from two to about 10 carbon atoms. The phosphonic acids for use in the invention include phosphonic and polyphosphonic acids having between from two to about 12 carbon atoms.

20 Suitable for use as the salt are alkali metals, especially sodium, potassium, and lithium, as well as ammonium and amine. In addition, mixtures of these cations can be employed. The amines used to form the salts can be represented by the formula N(R)₃ or N(R)₄⁺ wherein R is independently selected from hydrogen or a C₁-C₈ alkyl group, a C₆-C₁₄ aryl group or a C₇-C₁₆ alkaryl or alkaryl group.

25 It is not necessary that the carboxylic or phosphonic acid salt be completely neutralized to be effective for use in the invention since only one of the acid groups of a polycarboxylic acid need be in the salt form.

Representative examples of the aminopolycarboxylic acid salts include salts of ethylenediaminetetraacetic acid (EDTA), nitrilotriacetic acid (NTA), iminodiacetic acid (IDA), N-(2-hydroxyethyl)iminodiacetic acid (HIMDA), diethylenetriaminepentaacetic

acid ((DTPA), N-(2-hydroxyethyl) ethylenediaminetriacetic acid (EDTA-OH) and glycol ether diaminetetraacetic acid (GEDTA).

5 Examples of the aliphatic acid salts include salts of oxalic acid, succinic acid, glutaric acid, and pyruvic acid. Exemplary of the amino acid salts include salts of glycine, alanine and lysine. Examples of the hydroxycarboxylic acid salts are salts of malic acid, citric acid, glycolic acid, lactic acid and tartaric acid.

Examples of the phosphonic acid salts include salts of iminodimethylphosphonic acid (IDP), alkyldiphosphonic acids (ADPAs) and polyphosphonic acid.

10 The herbicidal composition for use in the invention may further contain a mixture of two or more of the carboxylic or phosphonic acid salts referenced herein.

15 The active herbicidal composition is prepared by dissolving the carboxylic or phosphonic acid salt in water. The herbicide salt solution can also be prepared by mixing the free acid in water and adding the required basic compound (for example, lithium hydroxide, sodium hydroxide, sodium carbonate, sodium bicarbonate, potassium hydroxide, ammonium hydroxide, amine or quaternary ammonium hydroxide) in a sufficient amount to neutralize one or more of the acid groups. The addition could also be reversed, that is, the acid may be added to a water mixture of the required amount of basic compound.

20 The undesirable vegetation is then treated by wetting the leaves with the resulting herbicidal composition. Death of the wetted undesirable plants occur within one day to one week depending upon ambient temperature and the growing season of the undesirable plants. The younger the plant, the more susceptible it is to be killed. Under certain conditions, more than one treatment may be required to obtain complete control.

25 Weeds and grasses which may be controlled by the method of this invention include oxalis, quakgrass, crabgrass, bermudagrass, nimblewill, and speedwell.

30 The amount of carboxylic or phosphonic acid salt in the herbicidal composition varies with the weed to be destroyed and generally ranges from about 0.5 to about 20 wt. percent. The most effective concentration of carboxylic or phosphonic acid salt in the herbicidal composition is between from 1 and about 10 wt. percent. The effectiveness of weed control with the herbicides of the invention also depends on the ambient

temperature at the time of treatment. The warmer the weather, the more effective is the desired control.

The amount of herbicidal composition applied to the targeted vegetation is generally between from about 25 to about 150 lbs. per acre.

5 A surfactant may be added to the solution to aid in wetting the leaves of the treated plants but is not necessary to achieve effective control of the undesired vegetation. Surfactants increase the wetting ability of the composition for plant foliage and facilitate the distribution of the composition to the foliage. Either ionic or non-ionic surfactants may be used. Illustrative of classes of stable surfactants are nonionics such as the 10 ethylene oxides condensates such as of alkylphenols or fatty alcohols and lignosulfonates, ammonium salts of mixtures of aromatic sulphone sulphonic acids condensed with formaldehyde; anionic surfactants such as 1-hydroxyethyl-2-heptadecenyl gloxalidin as well as amine oxide surfactant. The concentration of surfactant should be at least about 0.05, generally at least about 0.1, and preferably at least about 0.2 weight percent of the 15 herbicidal composition.

The composition for use in the invention may further contain other additives such as coloring agents, thickeners and the like.

Herbicidal compositions for use in the invention are non-toxic and biodegradable. For example, the salts of EDTA are widely used in household and industrial cleaners, as 20 antioxidants in food and in the treatment of heavy metal poisoning in both humans and animals. Disodium citrate is used in medicine as an anticoagulant to prevent the clotting of blood intended for transfusion.

The carboxylic or phosphonic acid salt for use in the invention is readily available commercially at low cost. Since the only solvent needed is water, the cost of the final 25 herbicidal solution is very low.

The following examples will illustrate the practice of the present invention in its preferred embodiments. Other embodiments within the scope of the claims herein will be apparent to one skilled in the art from consideration of the specification and practice of the invention as disclosed herein. It is intended that the specification, together with the

examples, be considered exemplary only, with the scope and spirit of the invention being indicated by the claims which follow.

Example 1

A 1% solution of tetrasodium ethylenediaminetetraacetate was prepared by dissolving 10 g. of the salt in 990 ml. of water. This was sprayed on clumps of oxalis, quakegrass, crabgrass and nimblewill growing in a lawn of primarily fescue grass. The oxalis shriveled up and turned brown within a day. The quakegrass turned a grayish color within two days and within five days was shriveled and dead. The nimblewill and crabgrass turned brown within two days and were dead after seven days. The daytime temperature at the time of spraying was about 85° F. None of the surrounding desirable grass was affected by the treatment nor did any of the treated weeds grow back.

Example 2

A similar experiment to Example 1 was run using 1% diammonium ethylenediaminetetraacetate. The results were the same. Subsequently, large patches of nimblewill in turf grass were treated with 1% diammonium EDTA salt solution. Within two weeks, all of the treated nimblewill was dead without any effect on the rest of the lawn.

Examples 3, 4 and 5

Water solutions of 3% sodium citrate, 3% potassium citrate and 3% trisodium nitrilotriacetate were prepared by dissolving 30 g. of each salt respectively in 970 ml. of water. Each solution was then sprayed on separate clumps of quakegrass. In each case the treated weeds shriveled and died within a week. No surrounding plants were affected.

Example 6

A solution of 2% tetrasodium EDTA was sprayed on burmudagrass growing along a sidewalk and in cracks in the sidewalk. Within five days the bermudagrass had turned brown and started to shrivel but some green was still present. The bermudagrass was retreated and in another week it was completely dead. None of the treated bermudagrass grew back nor were any of the other plants in the sprayed area affected.

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Example 7

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A 10% solution of diammonium glutarate was prepared by neutralizing 50 g. of glutaric acid in water with aqueous ammonium hydroxide and bringing the final solution to 500 ml. with water. This was sprayed on wild ivy and quakegrass. Both were dead within 36 hours.

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None of the solutions described in any of Examples 1-7 had any effect when sprayed on perennial ryegrass, azaleas, pachysandra, vinca minor, liriope, English ivy, fern, lily-of-the-valley, and other similar desirable plants. This demonstrates the selective nature of the herbicides of the invention.

Example 8

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A 5% solution of dimethylammonium glycolate was prepared by dissolving 25 g. Of anhydrous glycolic acid in 400 ml. Of water and then adding 40% dimethylamine in water until a pH of 7 was reached as indicated by pH paper. The solution was then made up to 500 ml. with more water and a few drops of Ivory conc. detergent added. The solution was then sprayed on lawn grass invaded by the weeds oxalis, quakegrass, nimblewill and speedwell. Within 24 hrs, the oxalis was brown and shriveled. The nimblewill and speedwell turned gray and appeared dried out. The quakegrass leaves were curled and also appeared dried out. In a week all of the weeds were brown and dried up and appeared dead. The grass was unaffected.

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Example 9

A 5% solution of dimethylammonium lactate was prepared as described in Example 7 using lactic acid instead of glycolic acid. This solution was sprayed on quakegrass in turf in one location and on nimblewill turf in another. Within a week, most of the quakegrass leaves had turned white but some of the plants were still green. The area was sprayed again with the same solution. In another week all of the quakegrass was dead. No other vegetation was affected. In the nimblewill plot, most of the sprayed weeds turned brown in a week but not all of the nimblewill was dead. Another spraying was made. In another week, the rest died.

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Example 10

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A patch of quakegrass growing among lily-of-the-valley, liriope, wild strawberries and fern was sprayed with a water solution of sodium gluconate. Within a week, the quakegrass leaves appeared burned on the edges but the plants were not dead. The plants were sprayed again. In several days, the burned appearance of the leaves increased. A third spraying was made which caused all of the quakegrass to turn brown and withered. None of the other vegetation was affected in any way.

Example 11

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A solution was prepared by dissolving 10 g. of diammonium ethylenediamine tetraacetic acid in 450 ml. of 50% isopropanol. This solution was then sprayed on crabgrass. In two days, the crabgrass turned brown and shriveled. In four days, it appeared to be completely dead. This demonstrates that not all of the acid groups in a complexing acid need to be in the salt form for the complexing compound to be an effective herbicide.

CLAIMS

What is claimed is:

1. A method for the control of undesired vegetation which comprises applying to the locus of the undesired vegetation a herbicidally effective amount of a composition comprising water and a salt of a carboxylic or phosphonic acid capable of forming a stable coordination compound with calcium or magnesium ions.
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2. The method of Claim 1, wherein the salt of the carboxylic or phosphonic acid is a salt of an acid selected from aminopolycarboxylic acids, aliphatic carboxylic acids, hydroxycarboxylic acids, amino acids, ether polycarboxylic acids, phosphonic acids and polyphosphonic acids.
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3. The method of Claim 2, wherein the alkali of the alkali salt is sodium, potassium, lithium, or ammonium.
4. The method of Claim 2, wherein the aminopolycarboxylic acid is ethylenediaminetetraacetic acid, nitrilotriacetic acid, iminodiacetic acid, N-(2-hydroxyethyl)iminodiacetic acid, diethylenetriaminepentaacetic acid, N-(2-hydroxyethyl)ethylenediaminetriacetic acid or glycol ether diaminetetraacetic acid.
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5. The method of Claim 2, wherein the aliphatic carboxylic acid is oxalic acid, succinic acid, glutaric acid or pyruvic acid.
6. The method of Claim 2, wherein the amino acid is glycine, alanine or lysine.
7. The method of Claim 2, wherein the phosphonic acid is iminodimethylphosphonic acid, alkyldiphosphonic acid or polyphosphonic acid.
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8. The method of Claim 2, wherein the hydroxycarboxylic acid is malic acid, citric acid, glycolic acid, lactic acid or tartaric acid.

9. The method of Claim 3, wherein the acid is ethylene diamine tetraacetic acid.

10. The method of Claim 3, wherein the acid is nitrilotriacetic acid.

11. The method of Claim 4, wherein the acid is glutaric acid.

12. The method of Claim 2, wherein the salt of the carboxylic or phosphonic acid is
5 a salt of the formula $N(R)_3$ or $N(R)_4^+$ wherein R is independently selected from hydrogen or a C_1 - C_8 alkyl group, a C_6 - C_{14} aryl group or a C_7 - C_{16} alkaryl or alkaryl group.

13. The method of Claim 2, wherein the undesired vegetation is oxalis, quakgrass, crabgrass, bermudagrass, nimblewill, or speedwell.

14. The method of Claim 2, wherein the composition further contains a surfactant.

10 15. The method of Claim 14, wherein the surfactant is non-ionic or anionic.

16. The method of Claim 16, wherein the surfactant is an ethylene oxide condensate, lignosulfonate, ammonium salt of mixtures of aromatic sulphone sulphonic acids condensed with formaldehyde, 1-hydroxyethyl-2-heptadecenyl gloxalidin or an amine oxide.

15 17. A method for the control of undesired vegetation which comprises applying to the locus of the undesired vegetation a herbicidally effective amount of an aqueous composition containing carboxylic or phosphonic acid salt which is the reaction product of:

20 (A) lithium hydroxide, sodium hydroxide, sodium carbonate, sodium bicarbonate, potassium hydroxide, ammonium hydroxide, amine or quaternary ammonium hydroxide; and

(B) oxalic acid, succinic acid, glutaric acid, pyruvic acid, glycine, alanine, lysine, malic acid, citric acid, glycolic acid, lactic acid, tartaric acid, iminodimethylphosphonic acid, alkyldiphosphonic acid or polyphosphonic acid.

18. The method of Claim 17, wherein the aqueous composition further comprises a
5 surfactant.

19. The method of Claim 17, wherein the weight percent of carboxylic or phosphonic acid salt in the aqueous composition is between from about 1 to about 10 weight percent.

20. The method of Claim 17, wherein the undesired vegetation is oxalis, quakegrass, crabgrass, bermudagrass, nimblewill, or speedwell.



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(57) Abstract					
<p>A method for selectively controlling undesirable vegetation by contacting the leaves with a herbicidal composition comprising a carboxylic or phosphonic acid salt known to form a coordination compound with calcium or magnesium salts. In a preferred embodiment, the carboxylic or phosphonic acid salt is an alkali salt of at least one acid selected from an aminopolycarboxylic acid, an aliphatic carboxylic acid, a hydroxycarboxylic acid, an amino acid, an ether polycarboxylic acid, a phosphonic acid and a polyphosphonic acid.</p>					

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INTERNATIONAL SEARCH REPORT

Internal ref Application No

PCT/US 99/16232

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 A01N37/04 A01N37/36 A01N37/44

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 A01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category ^o	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	<p>CHEMICAL ABSTRACTS, vol. 123, no. 21, 20 November 1995 (1995-11-20) Columbus, Ohio, US; abstract no. 278721, OORA, JUNKO ET AL: "Herbicides containing fatty acids and ethylenediaminetetraacetic acid salts and weed control with the herbicides" XP002126239 abstract & JP 07 215806 A (NIPPON TOBACCO SANGYO, JAPAN) 15 August 1995 (1995-08-15)</p> <p>---</p> <p>US 3 998 967 A (STAUFFER CHEMICAL COMPANY) 21 December 1976 (1976-12-21) example 3</p> <p>---</p> <p>---</p>	1-4,9, 12-16
X		1-4,10, 12-16

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

* Special categories of cited documents :

- *A* document defining the general state of the art which is not considered to be of particular relevance
- *E* earlier document but published on or after the international filing date
- *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
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- *&* document member of the same patent family

Date of the actual completion of the international search

20 December 1999

Date of mailing of the international search report

13. 04. 2000

Name and mailing address of the ISA

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INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 99/16232

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category ^a	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	<p>DATABASE WPI Derwent Publications Ltd., London, GB; AN 1995-212841 XP002126242</p> <p>JAPAN TOBACCO INC: "Herbicide which is safe to human body - contg. tropolone salt and/or Hinokitiol salt and EDTA salt" abstract & JP 07 126111 A</p> <p>---</p> <p>EP 0 760 207 A (KAO CORPORATION) 5 March 1997 (1997-03-05) * see the whole document*</p> <p>---</p> <p>CHEMICAL ABSTRACTS, vol. 89, no. 19, 6 November 1978 (1978-11-06) Columbus, Ohio, US; abstract no. 158688, TURNER, D. J. ET AL: "Complexing agents as herbicide additives" XP002126240 abstract & WEED RES. (1978), 18(4), 199-207,</p> <p>---</p> <p>CHEMICAL ABSTRACTS, vol. 85, no. 25, 20 December 1976 (1976-12-20) Columbus, Ohio, US; abstract no. 187626, ANON.: "Effect of di- and trivalent cations on the herbicidal activity of N-phosphonomethylglycine" XP002126241 abstract & RES. DISCL. (1976), 148, 10,</p> <p>-----</p>	1-4,9, 12-16
X		1-4,9, 10,12-16
X		1-4,9, 10,12-16
X		1-4,9, 10,12-16

INTERNATIONAL SEARCH REPORT

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Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. Claims Nos.: because they relate to subject matter not required to be searched by this Authority, namely:

2. Claims Nos.: because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:

3. Claims Nos.: because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.

2. As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:

4. No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

1-3, 12-16 (partially); 4, 9-10

Remark on Protest

The additional search fees were accompanied by the applicant's protest.

No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

1. Claims: 1-3(partially), 4,9-10, 12-16(partially)

A method for the control of undesired vegetation which comprises applying to the locus of the undesired vegetation a herbicidally effective amount of a composition comprising water and a salt of an aminopolycarboxylic acid

2. Claims: 1-3(partially), 5,11,12-20(partially)

A method for the control of undesired vegetation which comprises applying to the locus of the undesired vegetation a herbicidally effective amount of a composition comprising water and a salt of an aliphatic carboxylic acid

3. Claims: 1-3(partially), 8, 12-20(partially)

A method for the control of undesired vegetation which comprises applying to the locus of the undesired vegetation a herbicidally effective amount of a composition comprising water and a salt of a hydroxycarboxylic acid

4. Claims: 1-3(partially), 6, 12-20(partially)

A method for the control of undesired vegetation which comprises applying to the locus of the undesired vegetation a herbicidally effective amount of a composition comprising water and a salt of an amino acid

5. Claims: 1-3 (partially), 12-16 (partially)

A method for the control of undesired vegetation which comprises applying to the locus of the undesired vegetation a herbicidally effective amount of a composition comprising water and a salt of an ether polycarboxylic acid

6. Claims: 1-3(partially), 7, 12-20(partially)

A method for the control of undesired vegetation which comprises applying to the locus of the undesired vegetation a herbicidally effective amount of a composition comprising water and a salt of phosphonic or polyphosphonic acid

INTERNATIONAL SEARCH REPORT**II. Information on patent family members**

International Application No

PCT/US 99/16232

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
JP 7215806	A 15-08-1995	NONE	
US 3998967	A 21-12-1976	NONE	

